

# **Advanced Computational Methods for Magnetic Interactions in Metamagnetic Materials**

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First principles electronic structure techniques for the determination of magnetic properties have made tremendous progress, and when coupled with modern computational capabilities there is now the opportunity to bridge the gap between the atomistic length scale and the mesoscopic length scale where many magnetic properties of practical interest are determined. We propose to apply these novel electronic structure methods to magnetic metamaterials combining soft and hard magnetic materials at the nanoscale. A key characteristic of such materials is strong noncollinearity in the boundary region ("domain wall") between the hard and the soft component, which ultimately determines whether these materials will be fit for practical applications. We have developed a new approach to study these types of problems, which has been applied to propagation of domain walls and their interactions with defects in realistic magnetic materials (CoPt and FePd). The calculations require no parameters and can include the effects of time varying external fields and temperature, very important for magnetic metamaterials. For studying the latter, we will need to extend the method to allow the consideration of magnetic system with several hundreds atoms. For this purpose, we are currently combining the first principles theory developed at Ames with the non-collinear magnetic tight binding method developed at NRL. We also plan to add thermal heat baths to these simulations to study magnetic phase stability.

## **Introduction**

The dynamics of domain walls is a key issue that needs to be addressed to understand the physics of magnetization processes in high-performance

magnets. For instance, the interaction of domain walls with defects controls the coercive force. Thus understanding this interaction is a major component of new material design. To date, the theoretical description of spin dynamics in domain walls is rather qualitative, being based on simplified Heisenberg-like models. First principles calculations, in particular those performed at the Ames Lab, have shown that domain wall profiles in even the simplest magnets differ drastically from the tangential form predicted by the Heisenberg model. What severely limits the applicability of first principles calculations to such objects as domain walls is the relevant length scale of 10-50 nm (1000 atoms), while the relevant microphysics (exchange interactions and magnetic anisotropy) is characterized by the length scale of a few lattice parameters (0.1-1 nm). Coupling these length scales can be best handled by combining the first principle microscopic calculations with the highly accurate NRL-TB method.

This further links to micromagnetic models (up to microns) that incorporate anisotropy and exchange constants from *ab-initio* calculations. This combined first-principles TB-micromagnetic approach should be able to predict the existence and size of domain walls, describe their interaction with important defects (such as antiphase boundaries (APB), dislocations, etc), explore magnetotransport phenomena and produce key information about the nature of the coercive force and the shape of the hysteresis loop in technologically important bulk and multilayered materials. One of the main problems which such approach uniquely can address is a description of thermal properties when the parameters of Hamiltonian depend on temperature. We propose new Monte-Carlo –TB formalism to include such temperature dependence plus *ab-initio* dynamics included after thermalization is obtained.

## **First principle calculations (20-60 atoms)**

**Direct simulations of domain wall profile,  
critical field estimation,  
analysis of hysteresis loop**

**calculation of tight binding parameters  
calculation of exchange and anisotropy  
parameters**

## **tight binding method (100-1000 atoms)**

**NRL tight binding code + Monte-Carlo  
method =>  
reliable description of metamagnets at finite  
temperatures**

## **model simulations (1000000 atoms)**

**mean field method  
kinetic cluster method  
Monte-Carlo method**

## **Motivation**

A dynamics of domain wall in high-performance metamagnetic magnets is one of the key issues needed to understand the physics of coercive force in these magnets. The better we understand the interaction of domain walls with certain defects believed to influence the mechanism underlying the coercive force, the closer we are to systematic preparation of new materials based on theoretical predictions. Up to now, the theoretical description of spin dynamics in the domain wall with and without defects has been described using very crude Heisenberg-like of models without referring to a real system. A micromagnetic model, which incorporates anisotropy and exchange constants, can produce any results depending on values chosen for these constants. The first principles approach should be able to predict existence and size of domain walls, and describe their interaction with important defects (such as antiphase boundaries (APB), dislocations and so on).

## Significance

So far our studies have concentrated on obtaining pioneering theoretical results for a classical domain wall system, and on constructing accurate TB Hamiltonians for Fe and Ni. These computational tools will be combined for studying the interaction of domain walls and defects and should lead to a description of known high-performance hard magnets, and provide theoretical guidelines for the improvement of the coercive force and parameters of magnetization reversal in these materials. As a long-standing goal we would like to address is a study of thermal stability of magnetic properties. This stability is a key factor in a functioning of majority of magnetic devices (sensors, motors and etc) in the dangerous and extreme surroundings. The addition of real dynamics is also a unique study of the proposed work.

A unique feature of this collaboration is that the proposed Ames-NRL team has already developed both the theoretical formalism and the computational machinery to carry out this work. Such collaboration would be the only group having the above capabilities in place. It would be important to proceed with this project quickly. Both the theory developed at Ames and the TB works have been announced in recent meetings, which may enable other groups to catch up. It would be desirable to see Ames-NRL pioneering not only the original developments but to also accomplish the final goal of transferring the predictive power of first-principles calculations to the field of applied magnetism.

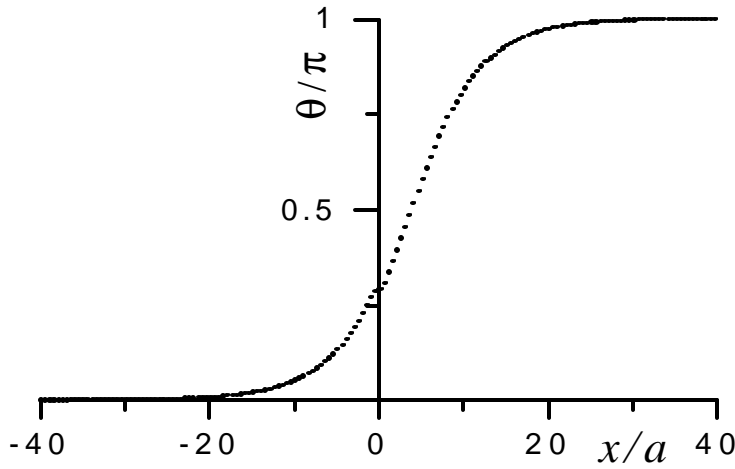
## Accomplishment

It was a common belief that electronic structure calculations are too time consuming even if a computer is capable of addressing a problem of such size (the domain walls are up to 1 micron in 3d alloys). Our estimations nevertheless reveal that many technologically important systems (such as FePt, CoPt and others) can be described in the framework of modern first principle calculations if proper optimization of the size of the system is made.

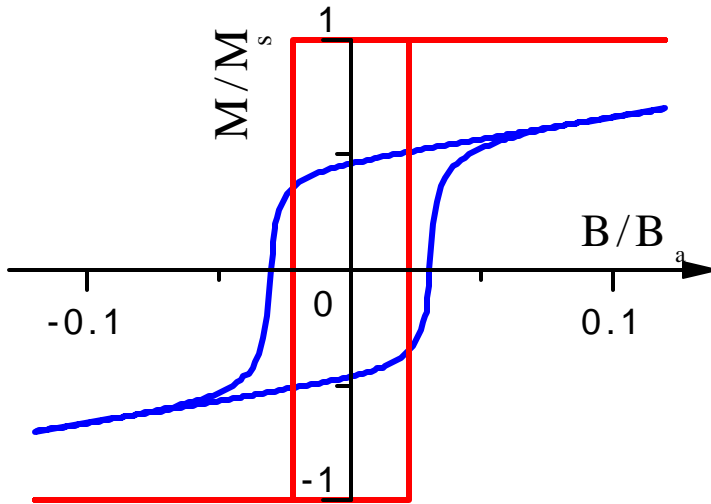
Using *ab-initio* linear muffin tin orbital method we first obtained the domain wall in the ideal CoPt system by direct optimization of magnetic structure between two domains. The size of the domain wall becomes stable after the corresponding size of the supercell exceeded approximately 100 atoms. Systematic increase of the supercell size has shown that the 180 degrees domain wall in ideal CoPt system along (100) with spin-orbit coupling pointing z direction is about 9 nm, with a structure generally similar to one predicted in Landau theory. Similar calculations for other materials

with similar AuCu structure have shown that the domain wall sizes in FePt and FePd should be about 14 nm and 18 nm , respectively.

We have also applied the scheme we developed to study the interaction of the obtained domain wall and APB defect (most common defect in this class of materials). The results obtained have shown that the (100) APB in CoPt repels the domain wall (Fig 1). In this case the domain wall avoids the region where the APB is localized, directly affecting the mechanism of coercive force formation in CoPt (Fig 2). A statistical analysis of different APB's and a study of domain wall dynamics for different microstructures in CoPt are in progress.



**Fig 1.** Profile of the domain wall with an antiphase boundary at  $x=0$ , for the external field  $B=B_c$  ( $B_c/B_a = 0.022$ ).



**Fig 2.** **Blue:** hysteresis loop of crystal CoPt after tempering for 30 min at 933 K. Applied field is along the (001) direction. **Red:** theoretical scheme for a single antiphase boundary parallel to the domain wall with exchange parameters taken from the *ab-initio* calculations.  $M_s$  is the saturation magnetization,  $B_a = 2K/M$  is the magnetic anisotropy field.